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PHASE TRANSITION KINETICS IN THE PRESENCE OF JOULEAN DISSIPATION

A. S. Pleshanov

A phase transition in the presence of volume heat liberation occurs in electrical breakdown of condensed media, in electrical explosion of conductors, and in various commutation processes. It was noted in [1, 2] that under conditions of volume heat liberation the phase transition includes a region of phase coexistence. However to the best of the author's knowledge the effect of volume heat liberation on the kinetics of the phase transition have yet to be studied. The present study will offer a theoretical analysis of this effect.

Let an electrical current of density j pass through a layer of condensed material. Due to Joulean dissipation the temperature T of the layer increases. The nonsteady-state T distribution can be found from the one-dimensional thermal conductivity equation

$$\rho w_t = (\varkappa T_x)_x + j^2/\sigma, \tag{1}$$

where $w = w^0 + cT$ is the enthalpy (w^0 is the reference level for measurement of w, c is the heat capacity); ρ is the density; $\varkappa$ is the thermal conductivity coefficient; σ is the electrical conductivity; x is the coordinate across the layer; the subscript denotes differentiation. The initial condition at time $t = 0$ has the form

$$T(x, 0) = T_0, \tag{2}$$

while the boundary condition corresponding to Newtonian heat exchange with the external medium at $T = T_0$ is

$$[\varkappa T_x + \alpha(T - T_0)]|_d = 0 \tag{3}$$

(where α is the heat exchange coefficient and d is the layer half-width). When T_{max} reaches the phase transition temperature $T_\ast$ a new phase appears, the process of heat propagation within which is described by an equation analogous to Eq. (1). On the phase boundary the conditions of continuity of j, and the mass and energy fluxes are satisfied, as well as continuity of the potential φ and T:

$$\{j\} = \{\rho v\} = \{\rho v w - \varkappa T_x\} = 0, \quad \{\varphi\} = \{T\} = 0. \tag{4}$$

Here v is the velocity relative to the seam and $\{f\} = f_2 - f_1$ (the subscripts $\alpha = 1, 2$ refer to the initial and new phases respectively). Sample φ and T distributions under conditions of identical heat exchange at both boundaries are shown in Fig. 1.

In fact, when T_{max} reaches the value $T_\ast$ under the conditions of the given problem, where heat is conducted to the layer volumewise and at a finite rate, a finite time is required for transition of phase 1 into phase 2. There follows from Eq. (1) an expression describing this nonsteady-state process at $T_1 = T_2 = T_\ast$:

$$\rho w_{\ast t} = j^2/\sigma_\ast, \tag{5}$$

where $w_\ast = \sum_\alpha w_{\alpha\ast} x_\alpha = w_{1\ast} + (w_{2\ast} - w_{1\ast}) x_2 \equiv w_{1\ast} + \Delta w_\ast x_2$; $\sigma_\ast = \sum_\alpha \sigma_{\alpha\ast} x_\alpha = \sigma_{1\ast} + (\sigma_{2\ast} - \sigma_{1\ast}) x_2 \equiv \sigma_{1\ast} +$

$\Delta\sigma_\ast x_2$; x_α is the volume concentration of the phase α, coinciding in view of the equality of the molecular weights of both phases with its mass fraction (it is assumed that σ_α is proportional to the volume density of charge carriers n); the subscript $\ast$ indicates $T = T_\ast$. Here we use an additive expression for the conductivity of the mixture, which gives a minimum value

for its resistance. The maximum resistance is obtained when an additive expression is used for the resistivities. The value of the actual resistance will lie between these limiting values. The integral of Eq. (5) for j = const has the form

$$\rho \Delta w_* \left(\sigma_{1*} x_2 + \Delta \sigma_* (1/2) x_2^2 \right) = j^2 (t - \theta)$$ (6)

(θ is an integration constant corresponding to the moment of appearance of phase 2 ($x_2 = 0$)); $\tau_1 = (t - \theta)_{max}$ is obtained from the condition of disappearance of phase 1 ($x_2 = 1$); $t_0 = \theta_{min}$ corresponds to the moment when the condition $T_1(0, t_0) = T_*$ is first achieved. At the time $t_1 = t_0 + \tau_1$ in the center of the layer phase 1 disappears completely, after which there departs from the center a front of abrupt transformation of phase 1 into phase 2 with decreasing value of the concentration x_{2*} on this front in the direction of the region of phase coexistence; x_{2*} can be found from Eq. (6) when the half-width of phase 2, equal to $\ell_2(t)$, is equal to the coordinate of the leading edge of the phase coexistence region at time θ, equal to $\ell_{12}(\theta)$. Moreover, using the symmetry boundary condition and the condition

$$T_{2,x}(0, t) = 0, \quad T_2|_{l_2} = T_{**}, \quad (x_2 T_{2,x} + \rho \Delta w_* x_{1*} l_{2,t})|_{l_2} = 0.$$ (7)

As regards determination of $\ell_{12}(t)$, the latter can be found from the temperature field in phase 1 for boundary conditions (3) and

$$T_1|_{l_{12}} = T_*, \quad T_{1,x}|_{l_{12}} = 0.$$ (8)

The last condition of Eq. (8) is a consequence of the absence of a gradient in the phase coexistence region. The rear edge of the phase coexistence region, defined by $\ell_2(t)$, overtakes the leading edge until finally the phase coexistence region disappears, after which the pattern of T distribution will coincide with Fig. 1b. Thus, between the stage of layer heating until the temperature $T_1(0, t)$ reaches the value T_* (stage 0) and the stage of nonsteady-state transition from phase 1 to phase 2 (stage 3) there exist a stage of formation of a phase coexistence region from the center of the layer to its periphery (stage 1) and a stage of simultaneous presence of phases 1 and 2, separated by an intermediate phase coexistence region (stage 2). A qualitative x—t diagram with x_2 references is shown in Fig. 2, while sample T distributions for various stages are shown in Fig. 3.

A mathematical description of the given nonsteady-state process with nonlinear boundary conditions at unknown phase boundaries $\ell_{12}(t)$ and $\ell_2(t)$ is quite complex even for the model situation of constancy of $\sigma_\alpha(\sigma_1 \neq \sigma_2)$ and $\rho_1 = \rho_2$, which will be assumed below. A simple, and as will be shown below, sufficiently accurate method of study is that of integral balances, in which Eq. (1) is satisfied in integral form

$$\int_{l_{12}}^{d} \rho_1 w_{1,t}\, dx = \varkappa_1 T_{1,x}|_{l_{12}}^{d} + j^2 \int_{l_{12}}^{d} \frac{dx}{\sigma_1};$$ (9)

$$\int_{0}^{l_2} \rho_2 w_{2,t}\, dx = \varkappa_2 T_{2,x}|_{0}^{l_2} + j^2 \int_{0}^{l_2} \frac{dx}{\sigma_2},$$ (10)

while the T_α profiles are taken parabolic in x

$$T_\alpha = a_\alpha + b_\alpha x + c_\alpha (1/2) x^2$$ (11)

with coefficients dependent on t and determined from the boundary conditions. It can easily be proved that the system (9), (10) together with all the boundary conditions is closed relative to the six coefficients of Eq. (11) and the two laws of motion for ℓ_{12} and ℓ_2. Use of a power approximation in Eq. (11) is justifiable from the fact that a solution in the form of Eq. (11) for constant coefficients ρc, $\varkappa$, and j^2/σ is a particular exact solution of Eq. (1), asymptotically valid as $t \to \infty$.

We will demonstrate the accuracy of the integral balance method using the self-similar problem which has an exact solution. Upon a layer of material at $T = T_*$ let there act an electrical voltage U = const, which falls only across the newly appearing new phase. Equation (1) has the form

$$\rho w_t = (\varkappa T_x)_x + \frac{1}{6} \left(\frac{U}{\int_0^l dx/\sigma} \right)^2$$ (12)
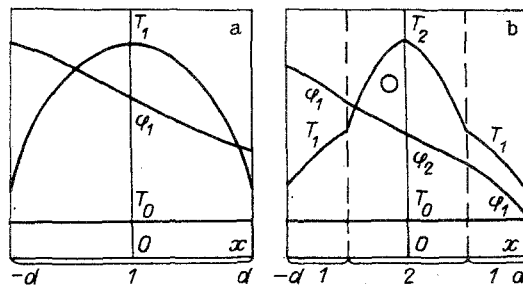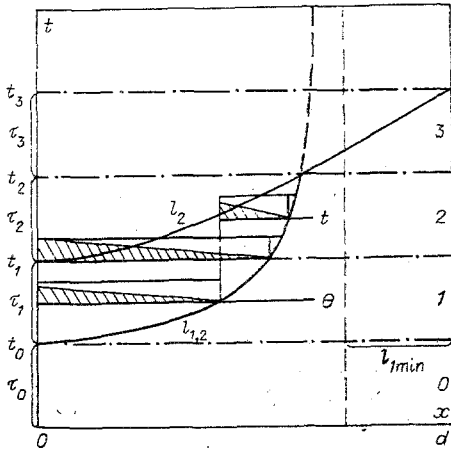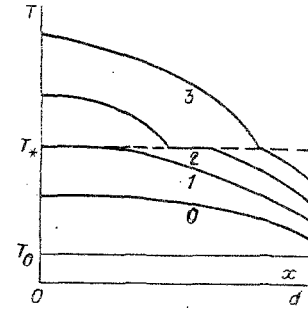
Fig. 1

Fig. 2

Fig. 3

(where ℓ is the width of the new phase). The temperature T satisfies the boundary conditions

$$T_x(0, t) = 0, \quad T|_l = T_*, \quad (\varkappa T_x + \rho \Delta w_* l_t)|_l = 0. \tag{13}$$

We will seek a solution of Eq. (12) in the form $T = T_* + (T_m - T_*)f(\xi)$, where $T_m = T(0, t) = $ const; $\xi = x/(2\sqrt{\chi t})$ being the self-similar variable. Obviously $f(0) = 1$ and $f(\eta) = 0$ ($\eta = \ell/(2\sqrt{\chi t})$). For ρc, $\varkappa = $ const

$$f_{\xi\xi} + 2\xi f_\xi + \frac{1}{\varkappa\sigma(T_m - T_*)}\left(\frac{U}{\int_0^\eta d\xi/\sigma}\right)^2 = 0,$$

while for $\sigma = $ const we have the exact solution

$$f = \left[\frac{U}{\sqrt{(\varkappa/\sigma)(T_m - T_*)}}\right]^2 \frac{1}{\eta^2}\int_\xi^\eta \exp\left(-\xi'^2\right)\left(\int_0^{\xi'}\exp\left(\xi''^2\right)d\xi''\right)d\xi',$$

which at $\xi = 0$ relates T_m and η; η is found according to Eq. (13) from

$$\eta\sqrt{\frac{\eta}{D(\eta)}} = \overline{U} \equiv \frac{U}{\sqrt{2(\varkappa/\sigma)(\Delta w_*/c)}} \tag{14}$$

(where $D(\eta) = \exp\left(-\eta^2\right)\int_0^\eta \exp\left(\xi^2\right)d\xi$ is a Dawson integral [3]). For this case the integral balance method gives the relationship

$$\overline{U} = \eta\sqrt{1 + (2/3)\eta^2}. \tag{15}$$

Figure 4 shows the exact η_1 and approximate η_2 solutions as functions of the parameter $\overline{U}$. The solutions begin to diverge at $\eta \gtrsim 1$, with the integral balance method giving the larger η value, i.e., more intense motion of the phase transition boundary. The reason for the good approximation can be demonstrated by power expansions of Eqs. (14), (15)

$$\overline{U} = \eta(1 + (1/3)\eta^2 + (1/30)\eta^4 - \ldots),$$
$$\overline{U} = \eta(1 + (1/3)\eta^2 - (1/18)\eta^4 + \ldots).$$

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We note that solution (15) is a particular solution of the corresponding nonlinear differential equation; the general solution of this equation does not satisfy the initial condition for arbitrary finite problem parameter values.

The accuracy of the integral balance method can also be tested by study of the layer heating state (stage 0) with the same assumption of constancy of all coefficients. The exact solution of Eq. (1) for the conditions $T_1(x, 0) = T_0$, $T_{1,x}|_0 = 0$, $[\varkappa_1 T_{1,x} + \alpha(T_1 - T_0)]|_\alpha = 0$, obtained in analogy to [4], has the form

$$T_1 - T_0 = 2 \frac{(jd)^2}{(\varkappa\sigma)_1} \sum_{n=1}^{\infty} \frac{\sin \lambda_n d}{(\lambda_n d)^3} \left(1 - \exp\left(-\lambda_n^2 \chi_1 t\right)\right) \cos \lambda_n x \equiv \tag{16}$$

$$\equiv \frac{(jd)^2}{(\varkappa\sigma)_1} \left\{ \left[\frac{1}{N_d} + \frac{1}{2}\left(1 - \frac{x^2}{d^2}\right) \right] - 2 \sum_{n=1}^{\infty} \frac{\sin \lambda_n d}{(\lambda_n d)^3} \exp\left(-\lambda_n^2 \chi_1 t\right) \cos \lambda_n x \right\}.$$

Here the eigenvalues λ_n satisfy the characteristic equations $\lambda_n d \tan \lambda_n d = N_d$; $\chi_1 = (\varkappa/\rho c)_1$ is the thermal diffusivity; $N_d = \alpha d/\varkappa_1$ is the Nusselt number; the term in square brackets is the steady-state solution of Eq. (1) as $t \to \infty$, if $T_{max} < T_\varkappa$. We write the solution of Eq. (1) bound by the integral balance method in the form

$$T_1 - T_0 = \frac{(jd)^2}{(\varkappa\sigma)_1} \left[\frac{1}{N_d} + \frac{1}{2}\left(1 - \frac{x^2}{d^2}\right) \right] (1 - \exp\left(-t/\tau_i\right)) \tag{17}$$

($\tau_t = (1/3 + 1/N_d)d^2/\chi_1$ is the thermal time).

Comparison of Eqs. (16) and (17) leads to the conclusion that the integral balance method is equivalent to the approximation of a regular regime where the first exponential in the series of Eq. (16) is considered. The moment when $T_1(0, t)$ reaches $T_\varkappa$ can be obtained from Eq. (17) in the form

$$t_0 \equiv \tau_0 = -\tau_t \ln [1 - (j_*/j)^2] \tag{18}$$

$\left(j_* = \frac{1}{\sqrt{1 + 2/N_d}} \frac{1}{d} \sqrt{2(\varkappa\sigma)_1(T_* - T_0)} < j \right.$ is the current scale), while the integral thermal balance equation at $t = t_0$ has the form

$$Q_{1,t}|_{t_0} \equiv (\rho c)_1 \int_0^d T_{1,t}|_{t_0} dx = \frac{d}{\sigma_1}\left(j^2 - j_*^2\right) > 0. \tag{19}$$

The following stage of formation of a phase coexistence region (stage 1) is described by the solution of Eq. (6), whence follows the duration of this stage

$$t_1 - t_0 \equiv \tau_1 = \frac{\rho \Delta w_*}{j^2} \frac{1}{2}(\sigma_1 + \sigma_2). \tag{20}$$

The coordinate of the leading edge of the phase coexistence region ℓ_{12} can be found by the integral balance method using the representation $\frac{T_1 - T_0}{T_* - T_0} = 1 - \frac{1}{1 + 2/N_1}\left(\frac{x - l_{12}}{l_1}\right)^2$, which satisfies boundary conditions (3) and (8). Here ℓ_1 is the width of phase 1 ($\ell_{12} + \ell_1 = d$); $N_1 = \alpha l_1/\varkappa_1$:

$$\frac{N_1(N_1 + 4)}{N_1 + 2} N_{1,t} = -\frac{3}{1/2 + 1/N_d} \frac{\chi_1}{d^2} [(j/j_*)^2 N_1(N_1 + 2) - N_d(N_d + 2)] \tag{21}$$

with initial condition $N_1(t_0) = N_d$. In analogy to Eq. (19)

$$\frac{N_d(N_d + 4)}{N_d + 2} N_{1,t}\bigg|_{t_0} = -\frac{3}{1/2 + 1/N_d} \frac{\chi_1}{d^2} N_d(N_d + 2)[(j/j_*)^2 - 1] < 0.$$

The integral of Eq. (21) has the form

$$\sum_{i=1}^{3} a_i \ln \frac{N_1 - N_{1i}}{N_d - N_{1i}} + \frac{3}{1/2 + 1/N_d} \frac{\chi_1}{d^2}\left(\frac{j}{j_*}\right)^2 (t - t_0) = 0, \tag{22}$$
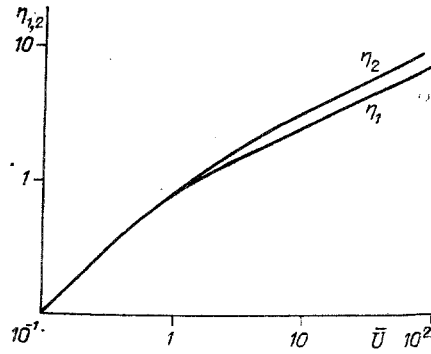
Fig. 4

where N_{1i} are the corresponding roots; a_i are quantities determined by the undefined coefficient method. For the special case $N_d \gg 1$ we obtain the simple integral

$$\ln \left[\frac{(j/j_*)^2 (l_1/d)^2 - 1}{(j/j_*)^2 - 1} \right] + 12 \left(\frac{j}{j_*} \right)^2 \frac{\chi_1}{d^2} (t - t_0) = 0, \tag{23}$$

whence it follows that $l_{1\min}/d = j_*/j < 1$. Analogously in the general case of Eq. (22) N_1 decreases from the value N_d to $N_{1\min} = \sqrt{1 + (j_*/j)^2 N_d (N_d + 2)} - 1 < N_d$. Thus the phase coexistence region does not extend beyond the layer boundary.

At the time $t = t_1$ a front of discontinuous transition into phase 2 begins to propagate from the layer center. The value of the phase 2 concentration on this front x_{2*} in the direction of the phase coexistence region can be obtained by eliminating θ from Eq. (6) and (22) or (23) (where we take $t = \theta$) with the condition $l_1 = d - l_2$. For example, with use of Eq. (23)

$$\frac{\rho \Delta w_*}{j^2} \left(\sigma_1 x_{2*} + \Delta \sigma \frac{1}{2} x_{2*}^2 \right) + \frac{1}{12} \left(\frac{j_*}{j} \right)^2 \frac{d^2}{\chi} \ln \left[\frac{(j/j_*)^2 - 1}{(j/j_*)^2 (l_1/d)^2 - 1} \right] = t - t_0 \tag{24}$$

$(t_1 \leq t \leq t_2)$, the extent of stage 2 can be found from the condition $x_{2*} = 0$ and is equal to

$$t_2 - t_1 \equiv \tau_2 = \frac{1}{12} \left(\frac{j_*}{j} \right)^2 \frac{d^2}{\chi_1} \ln \left[\frac{(j/j_*)^2 - 1}{(j/j_*)^2 (l_1/d)^2 - 1} \right] - \tau_1. \tag{25}$$

Commencing from the representation $T_2 - T_* = \frac{q_{2*}}{2 \chi_2 l_2} (l_2^2 - x^2)$, which satisfies boundary condition (7) ($q_{2*} = \rho \Delta w_* x_{1*} l_{2,t}$), we obtain the integral thermal balance equation

$$\frac{1}{3 \chi_2} (q_{2*} l_2^2)_t + q_{2*} = \frac{j^2}{\sigma_2} l_2, \tag{26}$$

from which, in view of the definition of q_{2*} with use of Eq. (24), we can find l_2. For small $t - t_1$ the velocity

$$l_{2,t} = 12 \frac{\chi_1}{d} \left[\left(\frac{j}{j_*} \right)^2 - 1 \right] > 0, \tag{27}$$

which proves constant and significantly higher than the thermal velocity χ_1/d.

The final stage 3 is described by Eq. (26) for phase 2 and an analogous equation for phase 1, which follows from the overall thermal balance

$$(\rho c)_1 \int_{l_2}^{d} T_{1,t} \, dx = \chi_1 T_{1,x} \big|_{l_2}^{d} + \frac{j^2}{\sigma_1} l_1 \tag{28}$$

and Eq. (11)

$$T_1 = T_* - \frac{q_{1*}}{\chi_1} (x - l_2) - \frac{1}{1 + 2/N_1} \left[(T_* - T_0) - \left(1 + \frac{1}{N_1} \right) \frac{q_{1*}}{\chi_1} l_1 \right] \left(\frac{x - l_2}{l_1} \right)^2, \tag{29}$$

which satisfies boundary condition (3) and

$$T_1 \big|_{l_2} = T_*, \quad -\chi_1 T_{1,x} \big|_{l_2} = q_{1*}. \tag{30}$$

The thermal fluxes introduced $q_{\alpha *}$ satisfy the boundary condition

$$q_{2*} - q_{1*} = \rho \Delta w_* l_{2,t}. \tag{31}$$

The system of Eqs. (26), (28) and (31) is closed with respect to $q_{\alpha *}$ and l_2.

Stage 3 is completed by exit of the phase transtiion boundary to the layer boundary (at time $t = t_3$), after which the process is described by a single Eq. (1) for the entire phase 2 layer until the onset of any following phase transition. Incidentally, second and subsequent phase transitions are possible before the first is completed.

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ROTATIONAL RELAXATION TIME OF NITROGEN

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Rotational relaxation can be described theoretically with the help of the Wang-Chang—Uhlenbeck equations for the one-particle distribution function $f_j(\mathbf{v}, \mathbf{r}, t)$ [1], where j is the rotational quantum number, $\mathbf{v}$ is the velocity, $\mathbf{r}$ is the position, and t is the time. When the charcteristic rotational relaxation time is much larger than the time to establish an equilibrium distribution of the translational energies of the molecules, the distribution function fj can be written in the form of a product fj = $f(\mathbf{r}, \mathbf{v}, t) N_j(\mathbf{r}, t)$. In this case relaxation of $N_j(\mathbf{r}, t)$ occurs for the equilibrium translational energy distribution and is described by the system of kinetic equations of [2]. Reliable information on the rate constants of the rotational transitions is not available at the present time, and this makes analysis of the rotational kinetics on the basis of these equations difficult.

A less detailed description can be obtained by using the relaxation equation

$$dE_R/dt = -(E_R - E_t)/\tau_R, \tag{1}$$

which is only valid when the deviation from equilibrium is small (E_R and E_t are the actual and equilibrium values of the rotational energy and τ_R is the rotational relaxation time).

The rotational relaxation time has been determined from various types of experiments (ultrasound, shock waves, thermal transpiration, and so on), including measurements of the parameters in a free jet [3]. It is usually assumed in analyzing the experimental results that (1) is valid over the entire flow field, although in free jets significant deviations from equilibrium can be reached, which makes the validity of this equation doubtful over the entire region of the parameters. A second deficiency of the analysis of existing experimental data is that the characteristic collision number $Z_R = \tau_R/\tau_t$ is assumed to be a constant over the entire flow field and is determined from certain quantities measured in the jet (τ_t is the translational relaxation time). However, it is well known that Z_R depends on the temperature of the gas, which varies significantly in the jet.

In the present paper the rotational relaxation time in molecular nitrogen is found in supersonic free jets using electron beam diagnostics. The population densities of the rota-